Coarse-graining molecular dynamics

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Outline

1. Motivation and Problem setup
2. General methodology
3. The derivation of the effective equations
   3.1 Static problems
   3.2 Dynamic problems
4. Implementations
Coarse-graining molecular dynamics

General framework

Motivation

- **System under consideration:** Solid systems with crystal structures.
- **Starting point:** Full molecular dynamics model with empirical potential
- **Observation:** Very often, the *interesting* atoms are in a localized domain.
- **Goal:** Deriving an effective model in which the effect of the *unimportant* atoms are included implicitly.
Example I: a single edge dislocation

- Sextic formulation of the anisotropic elastic field (Stroh 1958).
- Dislocation in a BCC system $\mathbf{b} = \frac{a_0}{2} < 111 >$.
- Atoms at the boundary are fixed.
Example II: an elliptical crack

- Anisotropic elastic solution (Sih and Liebowitz 1968).
- An open crack in a BCC system ($<110>$, $<1\bar{1}0>$, $<001>$).
- Atoms at the boundary are fixed.
Coarse-graining molecular dynamics

Main issues,
1. Selecting coarse-grain variables
2. Deriving effective equations
3. Efficient computational methods

Selecting coarse-grain variables near a crack.
Problem setup

As a boundary condition:
1. eliminating phonon reflection
2. model external loading
3. model heat bath to maintain system temperature

Partition of the atoms
Boundary condition for molecular statics

Partition of the displacement: \( \mathbf{u} = (\mathbf{u}_I, \mathbf{u}_J), \) \( \text{dim}(\mathbf{u}_I) \ll \text{dim}(\mathbf{u}_J) \)

- \( \mathbf{u}_I \): displacement of the atoms in the computational domain;
- \( \mathbf{u}_J \): displacement of the atoms in the bath.

Lattice Green’s function:

\[
- \sum_j D_{i-j} G_{j,k} = \delta_{ik}.
\]

Fourier representation of the Green’s function:

\[
G_j = \frac{1}{|B|} \int e^{i \mathbf{k} \cdot \mathbf{R}_j} D(\mathbf{k})^{-1} d\mathbf{k}.
\]
Formulation based on the Green’s functions

Linearize molecular statics model in the bath \( \sum_j D_j u_{i-j} = 0 \).

For any atom in the bath

\[
\begin{align*}
    u_n &= \sum_j \sum_k G_{j-k,n} D_k u_j \\
    &= \sum_j J_{n,j} u_j - \sum_i l_{n,i} u_i.
\end{align*}
\]

A more convenient form: (Li 2009)

\[
    u_J = \Theta u_I,
    \Theta = G_{JI} G_{IJ}^{-1}.
\]

The effective model

\[
    \min_{u_I} V(u_I, \Theta u_I).
\]
Coarse-graining molecular dynamics

Coarse-graining molecular statics

Example: an edge dislocation and an elliptic crack in Iron
Coarse-graining molecular statics

Defining coarse-grained variables: \( q = Bu \)
Eliminating the remaining degrees of freedom:

\[
\min V(u), \text{subject to } q = Bu.
\]

The mechanical equilibrium given: \( q, u = Rq, R = D^{-1}B^T(BD^{-1}B^T)^{-1}. \)

The effective model

\[
\min_q V(Rq).
\]
Projection formalism


Dynamics of the entire system:

\[ m\ddot{u}_i = -\nabla_{u_i} V. \]
Coarse-graining molecular dynamics

Coarse-graining molecular statics

Projection formalism


Dynamics of the entire system:

\[ m \ddot{u}_i = -\nabla u_i V. \]

Partition of the system:

- Retained variables
- Heat bath variables
Mori-Zwanzig’s formalism

1. Projection operator (conditional expectation),

\[ P g = E(g | \text{retained variables}), \quad Q = I - P. \]

2. For any function of the retained variables: \( \varphi \),

\[ \frac{d}{dt} \varphi(t) = e^{tL}L \varphi(0) = e^{tL}PL \varphi(0) + e^{tL}QL \varphi(0), \]

3. Dyson’s formula,

\[ e^{tL} = e^{tQL} + \int_0^t e^{(t-s)L}PL e^{sQL} ds. \]

4. Generalized Langevin equation:

\[ \frac{d}{dt} \varphi(t) = e^{tL}PL \varphi(0) + \int_0^t e^{(t-s)L}K(s)ds + R(t). \]

\[ R(t) = e^{tQL}QL \varphi(0), \quad K(t) = PLR(t). \]
Mori-Zwanzig’s formalism for Molecular Dynamics

Adelman and Doll 1974, 1976
Tully 1980
Sykes, 1976
Ciccotti and Ryckaert, 1981
Munakata, 1986
Izvekov and Voth 2006
Lange and Grubmüller, 2006
Darve, Solomon and Kia 2009
Mori-Zwanzig’s formalism for MD

Partition of the system:

$$u = (u_I, u_J), \quad v = (v_I, v_J)$$

The thermodynamic force:

$$e^{t\mathcal{L}\mathcal{P}\mathcal{L}v_I(0)} = -\frac{\partial W}{\partial u_I}.$$ 

The effective free energy:

$$W(u_I, T) = -k_B T \ln Z, \quad Z = \int e^{-\frac{V(u_I, u_J)}{k_B T}} \, du_J.$$ 

Coarse-graining MD based on the free energy (Rudd and Broughton 1998, 2005).
Coarse-graining molecular dynamics

Mori-Zwanzig’s formalism for Molecular Dynamics

Mori-Zwanzig’s formalism for MD – boundary conditions

Simplify the atomic interaction:

\[ V_{\text{new}} = V(u_I, B_{JI} u_I) + \frac{1}{2} (u_J - B_{JI} u_I)^T D_{JJ} (u_J - B_{JI} u_I). \]

Harmonic approximation:

1. it is only made for the atoms in the bath
2. the force constants are consistent with the atomic potential
3. serves as the first approximation of the original Mori-Zwanzig model

\[ R(t) : \text{stationary Gaussian process.} \]

\[ \left\langle R(t) R(s)^T \right\rangle = k_B T \Theta(t - s). \]
Mori-Zwanzig’s formalism for MD – the memory term

The memory term:

$$- \int_0^t \Theta(\tau) \dot{u}_I(t - \tau) d\tau.$$ 

The generalized Langevin equation: (Li and E 2007, Li 2008)

$$m \ddot{u}_I = -\nabla_{u_I} V - \int_0^t \Theta(\tau) \dot{u}_I(t - \tau) d\tau + R(t) + f_{\text{ex}}(t).$$
Example: molecular dynamics model for Iron
Example: fracture simulation

- anisotropic elastic solution (Sih and Liebowitz 1968).
- crack in a BCC system ($\langle 110 \rangle$, $\langle 1\overline{1}0 \rangle$, $\langle 001 \rangle$).
- generalized Langevin equations used at the boundary as the boundary condition.
Mori-Zwanzig’s formalism for MD – boundary conditions

Defining coarse-grained variables: \( q = Bu, p = Bv \)

Defining coarse-grained variables:
\[
P_u = D^{-1}B^T(BD^{-1}B^T)^{-1}B, \\
P_v = B^T(BB^T)^{-1}B
\]

The generalized Langevin equation:

\[
\begin{align*}
\dot{q} &= \frac{p}{m}, \\
M\dot{p} &= -R^T \nabla V(Rq) - \int_0^t \Theta(\tau)p(t - \tau)d\tau + R(t) + f_0
\end{align*}
\]
Example: One-dimensional chain

Choose one atom out of every $K$ atoms

Choose the average of every $K$ atoms
Exact and approximate memory kernels


Other related work:

- **nonlocal** in both space and time
- numerical implementation is expensive
- premature truncation leads to large reflection
Local memory kernels

(E and Huang, 2001; Li and E, 2006–2007)

- the memory kernel is independent of the temperature
- at zero temperature, similar to absorbing BC for wave equations
- minimize wave reflection

Basic principles:
1. Efficiency: local kernels
2. Stability: positive-definite kernels
3. Consistency: fluctuation-dissipation theorem
Variational approach

1. Express $\Theta(t)$ in the form of,

$$\Theta(t) = \int_{-\infty}^{+\infty} \Gamma(s)\Gamma(t + s)^T ds.$$  

$\Gamma(t)$ is local:

$$\Gamma_{ij}(t) = 0, \text{ if } |r_i - r_j| > r_c, \text{ or } |t| > t_c.$$  

2. Objective functions

$$\min_{\Gamma(t)} \int e(\omega; \Gamma) W(\omega) d\omega.$$  

3. Choose test functions,
   - Phonon mode
   - Green’s functions
Sample the random noise

The random noise $R(t)$ is a stationary Gaussian process. The fluctuation-dissipation theorem is satisfied:

$$\left\langle R(t)R(0)^T \right\rangle = k_B T \Theta(t).$$

Let $W(t)$ be white noise with variance $k_B TI$, then,

$$R(t) = \sum_k \int \Gamma(s)W(t - s)ds.$$
Case studied 1: fracture simulation

- anisotropic elastic solution (Sih and Liebowitz 1968).
- crack in a BCC system ($\langle 110 \rangle$, $\langle 1\bar{1}0 \rangle$, $\langle 001 \rangle$).
- generalized Langevin equations used at the boundary as the boundary condition.
Case studied 2: finite temperature crack simulation

At zero temperature
Case studied 2: finite temperature crack simulation

At finite temperature 500K
Summary

1. **Static problems**: Green’s function approach.
2. **Dynamic problems**: Mori-Zwanzig’s viewpoint
   Reducing the dimension of the problem
   ▶ linearized interaction bath variables
   ▶ thermal equilibrium initially for the atoms in the continuum region,
   ▶ projection operator using conditional expectation,
   ▶ generalized Langevin equations.
3. **Model the effective equations with GLEs**
   a. compute the memory kernels $\Theta$
   b. sample the random noise
   c. the fluctuation-dissipation theorem